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by

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16. ABSTRACT

A composite receptor model for PM-10 apportionment was evaluated to determine the stability of its solutions and to devise cost-effective measurement strategies. Ambient aerosol samples used in the evaluation were obtained with dichotomous samplers at three sites in the vicinity of Philadelphia, PA and were analyzed by x-ray fluorescence (XRF), instrumental neutron activation (INAA),scanning electron microscopy (SEM) and ion chromatography. The composite receptor model consisted of a wind trajectory method, chemical mass balances (CMB), and multiple linear regression (MLR). In resolving PM-10 into 11 components, SEM was essential for coal-fly ash and botanical matter, XRF was essential for a S-component and vehicle emissions, and INAA was essential for fluidized catalytic crackers at refineries. The remaining components were determined well by either INAA or XRF. Ten to twenty samples were sufficient to determine average source contributions by CMB. Multiple linear regression required 30 or more samples to determine the S-component and at least 50 samples for vehicle emissions.

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EVALUATION OF COMPOSITE RECEPTOR METHODS

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A composite receptor method for PM-10 apportionment was evaluated to determine the stability of its solutions and to devise cost-effective measurement strategies. Ambient aerosol samples used in the evaluation were obtained with dichotomous samplers at three sites in the vicinity of Philadelphia, PA and were analyzed by x-ray fluorescence (XRF), instrumental neutron activation (INAA) scanning electron microscopy (SEM) and ion chromatography. The composite receptor method consisted of a wind trajectory method, chemical mass balances (CMB), and multiple linear regression (MLR).

Several industrial sources were included in the CMB for fine particles, and vehicle emissions and a S-component were determined by MLR. Measured Pb minus a CMB-derived correction for other sources was used as an independent variable because vehicles contributed less than 80% of ambient Pb in a typical sample from Camden, NJ. MLR results for the Pb abundance in vehicle emissions agreed with predictions of a model for vehicle emissions.

In resolving PM-10 into 11 components, SEM was essential for coal-fly ash and botanical matter, XRF was essential for the S-component and vehicle emissions, and INAA was essential for fluidized catalytic crackers at refineries. The remaining components were determined well by either INAA or XRF. Ten to twenty samples were sufficient to determine average source contributions by CMB. Multiple linear regression required 30 or more samples to determine the S-component and at least 50 samples for vehicle emissions.

The standard dichotomous sampler collected fine particle as multiple layers that were unsuitable for analysis by SEM. A modified dichotomous sampler is described which prevents this problem and collects adequate samples for XRF, INAA and SEM analysis of both fine and coarse fractions.

Introduction

In a recent study a composite receptor method was applied to ambient aerosol data for 156 samples from three sites in the vicinity of Philadelphia, PA and Camden, NJ[1]. The composite method consisted of a wind trajectory method[2] to identify sources, chemical mass balance[3] (CMB) to determine their contributions to PM-10, and CMB combined with multiple linear regression (MLR) to determine a S component and vehicle emissions[1].

In developing the composite method, it was recognized that three criteria must be met for a species to be included in a CMB: (1) All sources of the species must be included in the CMB. (2) The element's abundance in particulate emissions from all of those sources must be known. (3) Sources with collinear signatures cannot be determined individually and must be included in a broadly defined category.

A EPA source signature library contains data for more than 120 sources[4], and many might be applicable to Philadelphia. However, collinearity becomes severe in a typical CMB if more than 6 to 12 components are included. Thus, the wind trajectory method[2] was valuable for selecting sources to include in CMB analyses. Components in CMBs for fine particles were crustal matter and emissions from municipal incinerators, oil-fired power plants, fluidized catalytic cracker, antimony roaster, and paint pigment manufacturing. Because signatures for coal-fly ash and soil were collinear, criterion 3 required that they be included in a broadly defined crustal category. Elements included in CMB analyses were Si, Ca, Ti, Ni, Zn, Cd, Sn and Sb measured by x-ray fluorescence (XRF) and Al, V, La, Ce and Sm measured by instrumental neutron activation analysis (INAA)[1].

Criterion 2 led to the exclusion of Pb and S from the CMB because their abundances were not known well for vehicle exhaust and a S-component. The latter consisted of sulfate, cations and possibly water or organic matter. Multiple linear regression (MLR) can be used to estimate S- and Pb-abundances but measured Pb cannot be used as an independent variable in MLR because vehicle exhaust was not the only significant source of Pb. Thus, the above described CMB was applied to each sample to determine non vehicular Pb, which was subtracted from measured Pb to yield a tracer for vehicle exhaust.

Scanning electron microscopy (SEM) provided estimates for several components including municipal incinerators[5]. For samples heavily impacted by municipal incinerators, SEM results confirmed the CMB results. However, cases were identified where CMB results indicated municipal incinerator emissions when none was detected by SEM[5]. Evaluation of those cases suggested possible departure from criterion 1 because Zn, an important tracer for incinerators, had other possible sources not included in the CMB. Thus, the incinerator component deduced by CMB is an upper limit, and results obtained by the MLR-CMB composite are potentially affected.

This work describes application of MLR to various data subsets chosen to reveal any dependence on Zn concentration, sampling site, sample size and measurement method. Apportionments presented here include SEM results for botanical matter and coal-fly ash in coarse particles[5].

Experimental Procedures

Aerosol samples were collected during consecutive 12-hour periods in dichotomous samplers at three sites between July 14 and August 13, 1982. Site locations are shown in Figure 1 and numbered 12, 28, and 34. Site 12 was at an airport in a residential, commercial area of northern Philadelphia.

Site 28 was in a commercial, industrial area of Camden, NJ. Site 34 was in a rural area near Clarksboro, NJ. All fine and coarse fractions samples were analyzed by β-gauge, ion chromatography (IC), and XRF. Only fine fraction samples were analyzed by INAA. A previous report demonstrated good agreement among IC, XRF and INAA results for $SO_4^=$, S, Mn, Fe, Zn and Sb[1]. Twelve coarse fraction samples were analyzed by SEM[5].

Analysis and Results

CMB Analysis of Philadelphia Aerosol

The CMB method expresses concentration C_i of species i as

$$C_i = \Sigma A_{ij} T_j \quad (1)$$

where A_{ij} is the abundance of species i in component j, and T_j is the total mass concentration of component j. This equation is solved for T_j by a least-squares method that uses effective-variance-weighting[3].

Five industrial sources[6] identified by wind-trajectory-analysis[1,2] and crustal matter[7] met the criteria outlined in the Introduction and were included in the CMB for fine particles. Table I shows results of CMB applied to the average composition of fine particles at site 28. The S-component and vehicle exhaust were excluded from the CMB and were determined by a composite of CMB and MLR.

Composite of CMB and MLR

Use of MLR in receptor models requires species that are unique tracers for sources. However, Table I shows that Pb is not a unique tracer for vehicle emissions because non vehicular sources contribute 22% of average Pb at site 28. In one sample municipal incinerators contributed 61% of ambient Pb. This problem is overcome by a composite method that uses CMB results to derive tracers M′, S′, and Pb′, which represent concentrations of mass, S and Pb due only to the S-component and vehicle emissions:

$$M' = M - \Sigma T_j \quad (2a)$$

$$S' = S - \Sigma A_{S,j} T_j \quad (2b)$$

$$Pb' = Pb - \Sigma A_{Pb,j} T_j \quad (2c)$$

Here M, S and Pb are the measured concentrations of mass, S and Pb. Data for each 12-hour sampling period were analyzed by CMB in the manner shown in Table I, and the resulting data were used in equation 2 to compute M′, S′ and Pb′, which were used in the following MLR:

$$M' = a + bS' + cPb' \quad (3)$$

A least-squares method was used to determine values of coefficients a, b and c that best represent the data. To identify any sensitivity to choice of data, equation 3 was applied to data for all 156 samples and to various data subsets, and results are shown in Table II. To assess the consequences of any large CMB error for the incinerator component, results are shown for Pb/Zn < 0.27. Such a constraint limits Pb from incinerators to less than 15% of ambient Pb; that constraint eliminates 46 out of 156 observations. Computations were also made for single sites. In other computations only every third or every fifth datum was used; results in Table II represent averages for various ways that every third or fifth datum can be selected.

Except for Pb results at site 34, regression results in Table II are not significantly affected by constraints on Zn, site or sample size.

Use of Br As a Tracer for Vehicle Exhaust

An advantage of Br as a tracer for vehicle exhaust is that Br has few other significant sources. A disadvantage is that Br is easily volatilized upon contact with acidic particles, and this makes it difficult to estimate Br′, the CMB-corrected parameter. Thus, Br, the measured bromine concentration was used in the following:

$$M' = a + bS' + dBr \qquad (4)$$

Results are shown in Table II both for all observations and for observations with Br/Zn < 6.8. Such a constraint limits Br from incinerator emissions to less than 15% of ambient Br. The constraint eliminates 8 out of 156 observations and has no significant effect on the results.

Abundances of S and Pb Derived from Regression Coefficients

For coefficients in equation 3, it is postulated that b^{-1} is the S abundance in the S-component, and c^{-1} is the Pb abundances in vehicle emissions. Intercept, a, represents additional sources uncorrelated with S and Pb. Abundances derived in this manner from the first row of results in Table II are 20.4 ± 0.4% for S in the S-component and 6.9 ± 1.5% for Pb in vehicle emissions.

Sulfur Abundance in S-Component. Sulfate in fine particles at site 28 was reported to be a mixture of ammonium sulfate and ammonium bisulfate[1]. The S-abundance is 24.2% in the former and 27.8% in the latter. The S abundance based on MLR is 16% and 27% lower than S abundances in ammonium sulfate and ammonium bisulfate, respectively. The discrepancy has two possible causes: (1) bias in measurement of mass or S and (2) water or carbon in the S-component.

A postulate of 16% measurement bias is not supported by results of method intercomparisons. For sites 12 and 28, mean XRF results for S were 7% lower than mean IC results for sulfate (expressed as S), and the correlation coefficient was 0.98. Furthermore, when sulfate (expressed as an equivalent amount of S) is used as a dependent variable in equation 3, the S abundance derived from the regression coefficient (shown in Table II) is 21 ± 1%, which is essentially the same as the result obtained by using S as an independent variable. (In these analyses, 3 out of 102 IC-sulfate values were outliers that were deleted.) In a comparison of β-gauge and gravimetric mass measurements in North Carolina, the ratio of means for 136 fine-fraction samples was 1.03, and the slope of a linear relationship was 1.03 ± 0.03[8].

Laboratory studies have demonstrated that ammonium sulfate and ammonium bisulfate aerosol particles can exist in hydrated states at relative humidity below the 55% RH of our mass measurements[9]. However, it is not known if such hydrated states can exist for particles on a filter. Also, the S-component could include organic carbon, but our carbon measurements lack the precision needed to test this possibility.

Pb Abundance in Vehicle Emissions. Emission rates and abundances of Pb in vehicle emissions were calculated from a model that uses comprehensive data for six classes of vehicles as a function of year of manufacture[10,11]. Computations were made assuming that local governments had programs to insure that emission systems were maintained. Total Pb emission rates and percent Pb in emissions weighted by vehicle-miles traveled (VMT) in Philadelphia

county[12] are plotted vs year in Figures 2 and 3. Figure 3 also shows results derived by MLR from ambient air measurements, which include the present result and results of other studies[13,14,15].

Model results for Pb abundance during 1982 in < 2.5-μm particles are 7.1% and 6.2% for the following extremes in driving conditions: 48-km/hr transient speeds and 88-km/h steady speed. Such results agree with 6.9 ± 1.5% deduced from the regression coefficient for Pb shown in the first row of Table II. Such agreement is encouraging, but sensitivity to input data makes results of the model uncertain. For example, the model assumes that 75% of Pb in fuel is emitted by vehicles, but measurements in a highway tunnel indicate only 50% is emitted[16]. Furthermore, the model only considers particles as they are emitted. When emissions dilute and cool in the atmosphere, volatile organic emissions can condense onto particles and cause the Pb abundance to decrease. Such considerations suggest that model results are uncertain by at least ±30%.

The model is also sensitive to the fraction of VMT due to each type of vehicle. Results described above are based on the following portions of VMT for heavy duty trucks in Philadelphia County: 5% gasoline and 6% diesel[12]. For traffic through a tunnel on the Pennsylvania Turnpike in western Pennsylvania, average VMT portions due to heavy duty trucks were 2.4% gasoline and 15.7% diesel[17]. Such turnpike data may better represent the vehicle component at site 34 because that site is only 1 km northwest of the New Jersey Turnpike. The model result, based on turnpike data for VMT and 88-km/h steady speed, is 2.6% Pb abundance, which agrees with the value, 3.8 ± 1.5%, deduced for site 34 from MLR results in Table II.

Concentrations for S-Component and Vehicle Exhaust

According to equations 3 and 4, a component's average concentration is the product of its regression coefficient and its mean tracer species concentration. Table III shows component concentrations derived in this manner using all data at all sites. Results based on Br and Pb tracers for vehicle emissions agree within estimated uncertainties. The Br results have the largest uncertainties, which may be due to the fact that Br can volatilize upon contact with acid sulfate particles. Thus, the most reliable regression results are based on Pb data. Results for the S-component and vehicle emissions at site 28 are shown in Table IV and are based on the S- and Pb-abundances derived by equation 3 for all 156 observations.

Sensitivity of Results to Choice of Measurement Method

Table IV shows apportionments based on the following combinations of measurement data: both XRF and INAA, only XRF and only INAA. When only XRF data were used, fluidized catalytic cracker emissions were not determined because essential elements (La, Se and Sm) were not detected. Except for that component and a marginally detected component from a paint pigment factory, the results are fairly insensitive to choice of measurement method.

Average uncertainties shown in Table IV are smaller for XRF results than for INAA results. This occurs because more XRF-measured elements were used in the CMBs. Fewer elements were used in the INAA based CMBs because INAA data were not available for the soil signatures in Philadelphia. This eliminated many elements such as Na, Sc, Cr and Co, which are accurately measured by INAA.

A comparison of measurement capabilities indicated that INAA data are superior for Na, Al, Se, V, Cr, Mn, Co, As, Se, Rb, Mo, Sb, I, Cs, La, Ce and Sm whereas XRF is superior for Si, S, Ca, Ni, Sr, Sn, and Pb[18]. Both methods

are comparable for K, Fe, Zn, and Br [18]. The present study generally confirms those rankings except that Mn and Sb concentrations were high enough to be accurately measured by XRF[1]. Such conclusions about INAA pertain to the National Bureau of Standards' reactor and do not necessarily apply to reactors with lower neutron flux or different energy distribution.

Sulfate measured by IC was not essential for apportionment of PM-10 and only served to confirm the accuracy of the XRF data for S. Nitrate data were of little value because of likely volatilization during sampling. Data on volatilizable carbon in this study had poor precision because sample blanks were large and variable.

Scanning Electron Microscopy

Individual particles in 12 coarse-fraction samples were identified by morphology and composition[5]. Morphology was obtained by SEM, and composition was obtained qualitatively by energy dispersive x-ray spectrometry (EDX). Many identified particles were characteristic of components determined by XRF data, but SEM-EDX provided unique data on two components: coal-fly ash and botanical particles[5]. Masses were estimated from observed dimensions and assumed densities[5], and results are shown in Table IV.

The standard dichotomous sampler collected fine particles as multiple layers that were unsuitable for analysis by SEM. The multiple layers were mostly sulfate, which coalesced due to deliquescence during high humidity, dried as large sulfate particles, and engulfed non sulfate fine particles[19]. Figure 4 shows a modified dichotomous sampler that collects fine particles on two separate filters. One collects at a low flow rate to avoid pileup. Such a sampler collects both fine and coarse particles in a manner compatible with requirements for analysis by SEM, XRF and INAA[19].

Conclusions

Measurement data were used to resolve PM-10 into 11 components. A composite of MLR and CMB was applied to XRF data, which was essential for a large S-component and vehicle emissions. INAA was essential for fluid catalytic crackers at refineries. SEM-EDX was essential for coal-fly ash and botanical matter. Remaining components were determined well by either INAA or XRF data.

Evaluation of data for 156 samples suggests a more economical and effective design for PM-10 apportionment in the Philadelphia study. Because INAA and SEM-EDX data were not used in MLR, it is only necessary to analyze enough samples to provide adequate estimates of the means. Standard errors computed from the standard deviations shown in Table IV suggest that 10 to 20 randomly selected samples analyzed by INAA and SEM-EDX should be sufficient. However, it is appropriate that all samples were analyzed by XRF because XRF analysis is less expensive and many samples analyzed by XRF were needed for the composite of CMB and MLR. Examination of data subsets indicated that the S-component was determined well by as few as 31 samples but the vehicle-component required more than 50 samples.

Analysis of sulfate collected in dichotomous samplers was of little value in the present study other than to confirm that XRF could be used to measure S in sulfate. In future studies, it would be more useful to collect samples in an annular denuder so that the nitrate volatilization problem can be avoided and so that both gaseous and particulate forms of S and N compounds can be analyzed[20].

In the Philadelphia study, two dichotomous samplers were used to collect samples for XRF and SEM analyses. The use of two dichotomous samplers is not only expensive, but the resulting fine fraction samples are unsuitable for SEM. The modified dichotomous sampler shown in Figure 4 provides a way to use one device to collect both fine and coarse particles that can be analyzed by XRF, INAA and SEM.

The abundance of S in the S-component derived by CMB-MLR was at least 16% lower than that of S in pure sulfate salts. Measurement error can account for no more than half of the difference. Much of the difference may be due to water or carbon bound to sulfate particles.

The abundance of Pb in vehicle emissions agreed with predictions of a comprehensive emissions model. If the model and its parameters were fully validated, it could be used instead of an expensive set of field measurements to derive the abundance of Pb in vehicle emissions. However, not all parameters in that model have been validated, and the model does not include particulate mass resulting from condensation of exhaust gasses in the atmosphere. Until the the model is validated for use with CMB, a composite of CMB and MLR will be needed to determine the Pb abundance.

The vehicle model predicts a factor of 20 decrease in Pb-emissions per unit of VMT between 1982 and 1990. Such a decrease implies that if the measurements shown in Table I were repeated in 1990, vehicles would contribute only 20% of ambient Pb. Such a low vehicle contribution will be a difficult challenge even for the composite of MLR and CMB. One way to overcome this difficulty in future studies would be to use CMB and MLR to deduce the Pb abundance in vehicle emissions sampled near a roadway. It is important that the roadway sampling site represents the vehicle mix and driving speeds of areas where PM-10 is to be apportioned. An alternative would be to use another tracer such as Br or CO. Br may be a poor tracer due to the tendency of Br to volatilize in the presence of acid aerosol. Carbon monoxide may be a better tracer for vehicles. Equivalence between MLR results based on CO and Pb was demonstrated for vehicles in Denver during 1982[15].

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Table I. Fine-particle CMB for site 28 based on XRF and INAA data[a].

Species	Meas. Meth.	Meas-ured	Calculated	Dust	Oil	Paint pig.	Catal. crack.	Incin-erator	Sb roast
In CMB									
Al	INAA	53	42 ± 14	20	5	2	14	2	0
Si	XRF	103	142 ± 40	77	16	0	36	12	0
Ca	XRF	40	38 ± 10	11	21	4	0	2	0
Ti	XRF	15	18 ± 9	3	1	13	1	0	0
V	INAA	13	11 ± 3	0	11	0	0	0	0
Fe	XRF	91	78 ± 26	55	12	9	0	2	0
Ni	XRF	11	13 ± 3	0	12	0	0	0	0
Zn	XRF	82	82 ± 7	1	7	0	0	74	0
Cd	XRF	2	1 ± 1	0	0	0	0	1	0
Sn	XRF	5	5 ± 4	0	0	0	0	5	0
Sb	XRF	79	79 ± 9	0	1	0	0	1	78
La*	INAA	1120	888 ± 201	16	208	0	662	1	0
Ce*	INAA	609	745 ± 182	42	162	0	542	0	0
Sm*	INAA	40	49 ± 15	6	9	0	34	0	0
Not in CMB									
S	XRF	4196	135 ± 287	1	86	0	25	21	1
Pb	XRF	249	55 ± 18	3	10	0	0	41	0
Mass	β	28282	2700 ± 3133	911	718	58	201	712	99

[a]Asterisk (*) indicates data in pg/m^3; all other data are in ng/m^3.

Table II. Multiple linear regression results and numbers of samples N for various choices of species, constraints, and measurement method.

N	constraint	Regression results[a],	R^2
XRF			
156	none	M′ = 3.6±0.8 + (14.0±3.2)Pb′ + (4.9±0.1)S′	0.91
110	Zn/Pb < 0.27[b]	M′ = 2.4±1.0 + (13.9±3.4)Pb′ + (5.1±0.1)S′	0.92
52	site 12	M′ = 3.8±1.4 + (9.2±5.4)Pb′ + (5.0±0.2)S′	0.91
50	site 28	M′ = 1.3±1.2 + (19.1±4.3)Pb′ + (5.2±0.2)S′	0.94
54	site 34	M′ = 3.8±1.7 + (26 ±10)Pb′ + (4.6±0.3)S′	0.87
52	every 3rd obs	M′ = 3.3±1.4 + (16.0±5.9)Pb′ + (4.9±0.2)S′	0.90
31	every 5th obs	M′ = 3.2±1.8 + (15.3±7.7)Pb′ + (5.0±0.3)S′	0.91
156	none	M′ = 3.8±0.8 + (71 ±20)Br + (5.0±0.1)S′	0.90
148	Zn/Br < 6.8[b]	M′ = 4.1±0.8 + (74 ±21)Br + (5.0±0.1)S′	0.90
XRF, IC[c]			
99	sites 12 & 28	M′ = 4.3±1.1 + (15.1±4.2)Pb′ + (4.8±0.2)$SO_4^=$	0.89
INAA			
155	none	M′ = 7.8±1.4 + (171±44)Br′ + (4.7±0.2)S′	0.72
140	Zn/Br < 6.8[b]	M′ = 8.0±1.4 + (168±45)Br + (4.6±0.2)S′	0.72

[a]Intercept has units of μg/m^3.

[b]This criterion limits Pb from incinerator emissions to < 15% of measured Pb.

[c]$SO_4^=$ is expressed as S, 3 outliers are deleted, and no data exist for site 34.

Table III. Concentrations in $\mu g/m^3$ for S-component and vehicle exhaust component based on MLR results in Table II.

Measurement Method	N	Variables	S-component	Vehicle emissions
XRF	156	M′ vs S′ and Pb′	19.2 ± 0.5	2.2 ± 0.5
XRF	156	M′ vs S′ and Br	19.6 ± 0.5	1.6 ± 0.4
INAA	155	M′ vs S′ and Br	16.5 ± 0.8	2.9 ± 0.7

Table IV. Comparison of average results in $\mu g/m^3$ at site 28 by composite receptor method based on various combinations of XRF, INAA and SEM-EDX data.

Component	XRF-INAA[a] SEM-EDX	XRF[b]	INAA[c]	standard deviation
Fine				
S-component	19.2 ± 0.5			
Vehicle exhaust	2.2 ± 0.5			
Regression intercept	3.5 ± 0.8			
Dust	1.0 ± 0.3	1.0 ± 0.3	1.4 ± 0.6	0.5
Oil-fly ash	0.7 ± 0.12	0.7 ± 0.14	0.8 ± 0.28	0.4
Paint pigment	0.05 ± 0.04	0.05 ± 0.04	na	0.08
Catalytic cracker	0.18 ± 0.04	na	0.17 ± 0.05	0.2
Incinerator	0.7 ± 0.1	0.7 ± 0.1	0.8 ± 0.1	1.5
Sb-roast	0.09 ± 0.01	0.09 ± 0.01	0.10 ± 0.01	0.2
Coarse				
Soil	d	6.7 ± 0.8		3.5
Sea salt	d	0.2 ± 0.2		0.6
Sulfate	d	0.5 ± 0.4		0.4
Incinerator	d	0.06 ± 0.2		0.2
Vehicle exhaust	d	0.7 ± 0.3		0.4
Sb-roast	d	0.23 ± 0.05		0.5
Coal-fly ash	0.2 ± 0.1			0.2
Botanical matter	1.8 ± 0.7			1.2
PM-10				
Calculated total	38.0 ± 1.7			
Measured total	40.1 ± 4.1	40.1 ± 4.1		

[a]Elements used in least-squares fit are shown in Table I.

[b]Based on fine data for Al, Si, S, Ca, Ti, V, Fe, Ni, Zn, Cd, Sn, Sb, and Pb and coarse data for Al, Si, S, Cl, Ti, Fe, Zn, Cd, Sb, and Pb by XRF.

[c]Based on INAA data for Al, Ca, V, Fe, Zn, Cd, Sb, La, Ce, and Sm.

[d]Same as XRF in next column because INAA was not applied to coarse fraction.

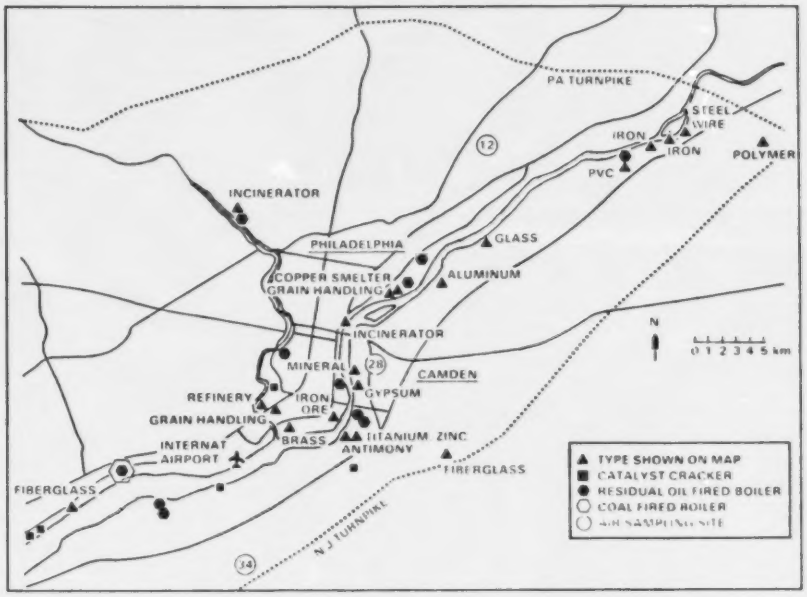

Figure 1. Map showing locations of sampling sites and major emission sources.

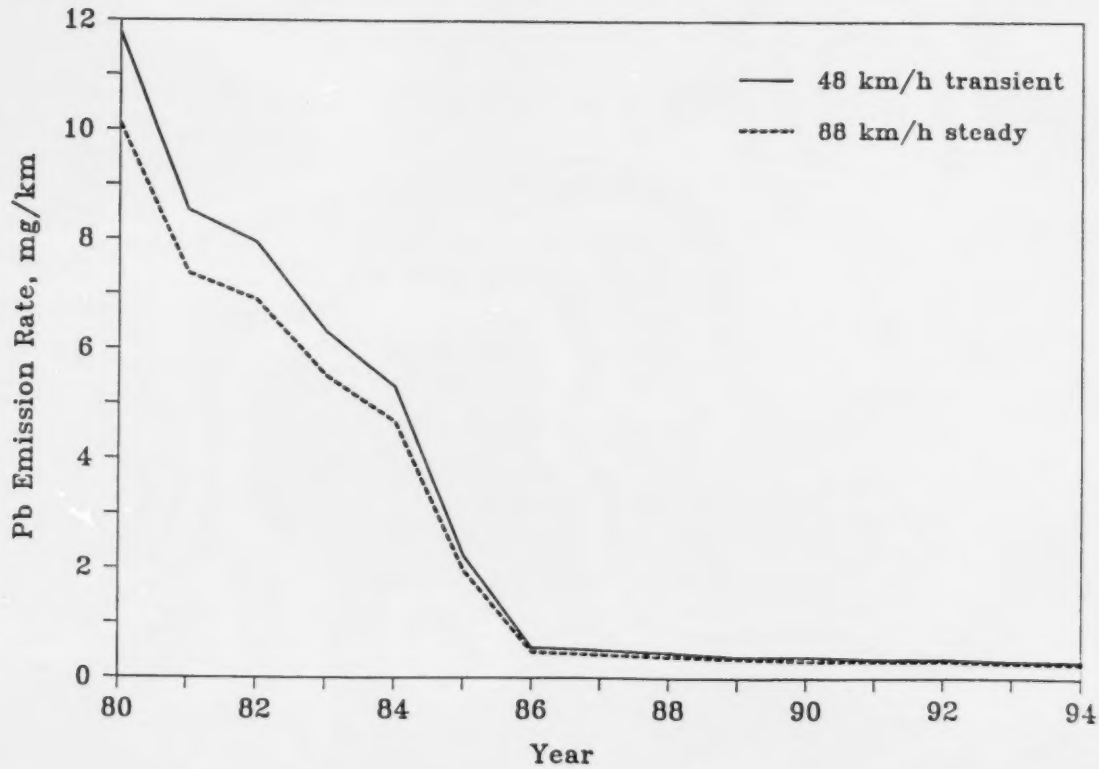

Figure 2. Model results for VMT-weighted Pb emission rate for < 2.5-μm particles from vehicles.

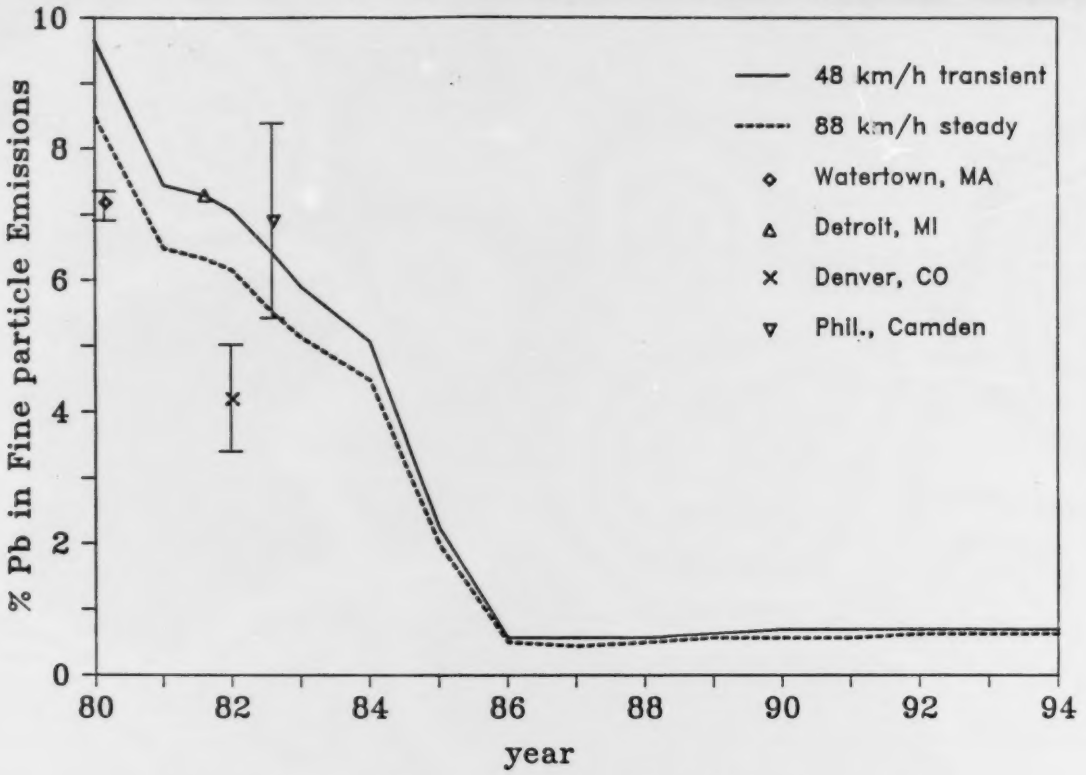

Figure 3. Model results for Pb abundance in < 2.5-μm particle emissions from population of vehicles representative of Philadelphia county. Data points represent MLR results applied to ambient aerosol data.

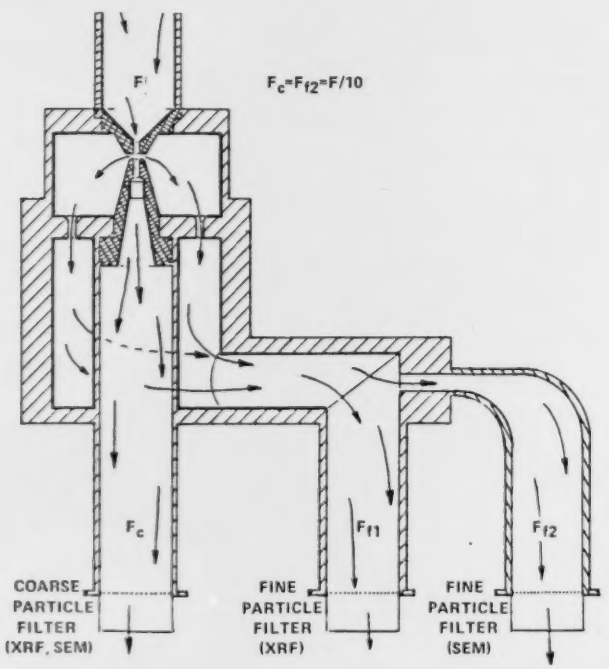

Figure 4. Modified dichotomous sampler that enables samples to be collected for analysis of individual particles and bulk composition.